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Functional Bounding Content Envelope for Actinides-Impact of Subcritical Multiplication

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ABSTRACT

The contents approved for shipment in a Type B radioactive material transportation package have historically been descriptions of discrete items, or groupings of well-defined similar items, in the package safety basis documentation. The need for a comprehensive functional content envelope of both gamma and neutron emitting nuclides compliant with regulatory limits has become necessary as the DOE complex requires shipments of unique mixtures of radionuclides and impurities. Recent publications have presented a calculational model and a corresponding content envelope intended to be compliant with federal regulatory external radiation limits as well as design decay heat limits based on the Model 9977 Packaging. The methodology used to develop this content envelope consisted of determining the external radiation dose rates based on one gram of a given isotope combined, in the case of actinides, with various levels of light element impurities and determining the allowable mass to the regulatory limits based on these dose rates. The method of ratioing up the masses to meet the regulatory limits fails in the case of some combinations of actinides and light element impurities because of the effect of subcritical multiplication. This is particularly true for many actinides combined with beryllium, boron, fluorine, lithium, and sodium. This paper rectifies this deficiency by adjusting the previously determined mass limits so that the resulting dose rates are compliant with regulatory limits. Some amount of iteration was required in some instances to adjust the masses for dose compliance. This paper will present a revised set of mass limits (i.e., content envelope) for several actinides with varying levels of light element impurities compliant with both external radiation and design decay heat limits. The revised content envelope covering a wide range of materials present in the DOE complex, was developed for the Model 9977 Packaging, providing conservative limits for other commonly-used Type B Packagings such as the Models 9975 and 9978.

INTRODUCTION

The specific contents approved for shipment in a Type B radioactive material transportation package have historically been descriptions of discrete items, or groupings of well-defined similar items, in the package safety basis documentation [i.e., Safety Analysis Report for Packaging (SARP)]. As new radioactive items were identified that needed to be shipped, an addendum to the safety basis was needed to add these new items. To develop, review and approve numerous addenda over time as more contents need to be added to the safety basis is both expensive and time consuming. Of late, there has been an increasing need at the DOE/NNSA complex to transport a wide range of radioactive materials, including sources and special nuclear materials and other actinides with light element impurities. A recent DOE report [1] jointly developed by three national laboratories provided a set of masses of both gamma and neutron emitting radioactive isotopes for the Model 9977 Packaging that were intended to be compliant with the external radiation limits set forth in 10 CFR Part 71 [2]. This set of masses also included mixtures of alpha-emitting actinides with beryllium impurities up to approximately 50% by mass of the mixture. This content envelope was further extended to include several light element impurities ranging

from 0.1% (1000ppm) to 90% by weight of the mixture [3]. The light-element impurities included were beryllium, boron, lithium, fluorine, carbon, aluminum, magnesium, sodium, silicon, oxygen, and chlorine. In addition to compliance with regulatory external radiation limits, the masses were also restricted by design limits on decay heat. However, it turned out that the methodology used to develop these limits tended to produce non-compliant external radiation levels for some combinations of actinides and light element impurities. On the other hand, there were several instances where the mass limits of specific isotopes in combination with light element impurities were overly conservative. This study was undertaken to rectify this problem and develop content envelopes that are compliant with regulations. The Model 9977 Packaging was used in this study to be consistent with the earlier studies [1, 3]. The limits presented for the Model 9977 Packaging are conservative for the Models 9975 and 9978 Packagings and specific analyses for these can be performed to remove this conservatism.

A DOE report [1] presented details of the methodology previously used to develop the bounding masses for both gamma and neutron emitters. The set of isotopes in this DOE report was from an earlier study [4] which presented a set of gamma sources and actinides developed from input from several DOE sources. In this earlier study [1], source spectra were established based on a 47-group structure for the neutron emitters and a 77-group structure for the gamma emitters. The source terms were generated using the source generating codes RASTA [5] or ORIGEN [6], depending on which one of these codes was suitable and bounding for a specific isotope. All source spectra were based on 1g of the isotope. The radiation transport calculations were performed using the Monte Carlo code, MCNP [7]. A detailed MCNP model of the Model 9977 Packaging was created and dose rates were calculated on the surfaces of the package as well as at 1 m from the surfaces as prescribed in 10 CFR Part 71 for non-exclusive use shipments. The dose rates were calculated as transfer functions starting with a single particle in each energy group for both the neutron and gamma source spectrum. The final dose rate at each point of interest was then estimated by folding in the appropriate source spectrum with the transfer functions. The sources were modeled as small cylinders (radius of 1.25 cm and height of 4 cm) placed at the bottom center of the containment vessel for the unshielded cases and at the bottom of the shielded container cavity for the shielded cases. The sources were modeled as voids as well as with a reduced density (1.27 g/cc) plutonium oxide to examine self-shielding and minimal subcritical multiplication effects of the source. The differences in the dose rates between the two sets were found to be insignificant.

However, when the masses determined from this previous study were modeled as spheres at the appropriate densities to maximize the effect of subcritical multiplication and located at the bottom of the containment vessel, the surface dose rate exceeded the regulatory limits for some actinides and actinide-light element mixtures. This was particularly true for higher concentrations of impurities such as beryllium, boron, fluorine, and lithium. For some other light-element impurities that are not strong (α, n) sources, the dose rates were actually well below the regulatory limits, especially at lower impurity concentrations. Thus, for a specific actinide and a light element impurity concentration, the impact on the dose rate is predicated on whether subcritical multiplication or self-shielding is the dominant factor. While the source terms were calculated based on the mixture of the actinide and light element, the source model used in the calculations assumed that it was entirely made of the actinide, thus increasing the probability of subcritical multiplication. The following sections will present an updated set of mass limits for the various actinides homogeneously combined with the light elements. In addition, the pure actinide mass limits were also recalculated to be compliant with regulatory limits. In all cases, design decay heat limits were also considered in defining the content envelope. Additionally, in the case of ^{239}Pu, masses that were in excess of 5000 g were restricted to this subcritical mass limit. The content

envelope is derived based on the Model 9977 Packaging with no shielded containers and will thus conservatively bound any shipments that use shielded containers as well as the Models 9975 and 9978 Packagings. It must be noted that criticality safety analyses prescribed by 10 CFR Part 71 were not performed for this content envelope and this aspect could further reduce the mass limits presented, even in the cases where the ^{239}Pu mass limit was set to 5000 g. The actinides studied included ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu+^{241}Am, ^{242}Pu, ^{244}Cm, ^{243}Am, ^{237}Np, ^{248}Cm, and ^{252}Cf. The last two are spontaneous fission neutron sources and the effect of light element impurities is negligible. Since ^{241}Pu has a short half-life and beta decays to ^{241}Am, the mass limit is given as the total of the two isotopes. While the allowable external radiation dose rate on the package surface (the limiting dose rate) for a non-exclusive use shipment is 200 mrem/h, the masses presented here are based on a limit of between 185 and 195 mrem/h, thus introducing an additional 7.5% to 2.5% conservatism to the content envelope.

MASS LIMITS FOR PURE ACTINIDES

Mass limits were recalculated for actinides without impurities. Table 1 presents the mass limits for the ten actinides. The mass limits for the two curium and the californium isotopes were reduced by approximately 33% compared to the mass limits presented in Reference 3. ^{238}Pu and ^{241}Pu+^{241}Am are limited by the design decay heat limit of 19W for unshielded contents in the Model 9977 Packaging. The ^{239}Pu mass limit has been set at 5000 g and could be further reduced because of criticality concerns. The two americium isotopes and neptunium showed large increases in the allowed mass compared those developed earlier [3], with ^{243}Am and ^{237}Np being predominantly gamma emitters. The mass limits shaded in orange represent limitations due to the 19 W decay heat limit for the contents without additional shielded containers.

Table 1. Mass Limits in grams for Pure Actinides

^{239}Pu	5000.0
^{238}Pu	33.46
^{240}Pu	2687.03
^{242}Pu	2003.19
^{241}Am+^{241}Pu	165.94
^{243}Am	1.32
^{237}Np	2982.00
^{244}Cm	0.26
^{248}Cm	0.07
^{252}Cf	0.0000012

MASS LIMITS FOR ACTINIDES WITH LIGHT ELEMENT IMPURITIES

^{238}Pu

The mass limits for this isotope were restricted by the design decay heat limits for several of the light elements that have low (α, n) cross sections. In the case of C, O, and Si, decay heat considerations limited the mass over the entire range of impurity levels that were studied. Even in the case of the typically strong (α, n) producing isotopes B, F, and Li, decay heat limits dictated the allowable mass at very low concentrations of these impurities. Table 2 presents the content envelope for ^{238}Pu.

Table 2. ^{238}Pu Mass Limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	14.13	33.46	33.46	33.46	33.46	33.46	33.46	33.46	33.46	33.46	33.46
5000ppm	2.99	11.88	33.46	30.99	33.46	33.46	33.46	33.46	33.46	33.46	33.46
10000ppm	1.49	6.10	33.46	15.98	33.46	33.46	33.46	33.46	33.46	33.46	33.46
50000ppm	0.33	1.35	14.99	3.50	33.46	33.46	24.63	19.66	33.46	33.46	33.46
10pc	0.18	0.75	8.48	1.91	33.46	31.25	13.55	10.87	33.46	33.46	33.46
30pc	0.08	0.35	3.95	0.84	33.46	16.62	6.02	4.80	33.46	33.46	33.46
50pc	0.06	0.27	3.05	0.63	33.46	10.23	4.50	3.63	33.46	33.46	33.46
70pc	0.06	0.23	2.66	0.54	33.46	8.70	3.84	3.04	33.46	33.46	33.46
90pc	0.05	0.21	2.44	0.48	33.46	7.83	3.46	2.75	33.46	33.46	30.90

^{239}Pu

In the case of ^{239}Pu combined with beryllium all combinations of the two except 1000ppm had to be reduced for compliance. Boron and fluorine mixtures up to a lower limit of 5% were reduced for compliance. For light elements with low (α, n) cross sections, the mass limits were restricted to its subcritical mass limit of 5000 g. In these cases the dose rates will have large margins to the regulatory limits. Table 3 presents the mass limits for ^{239}Pu.

Table 3. ^{239}Pu Mass Limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	2321.3	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0
5000ppm	900.0	2774.2	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0
10000ppm	476.0	1649.2	5000.0	3589.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0	5000.0
50000ppm	111.0	417.7	3629.0	1127.4	5000.0	5000.0	5000.0	4409.0	5000.0	5000.0	5000.0
10pc	62.5	240.0	2618.0	658.8	5000.0	5000.0	5000.0	3133.0	5000.0	5000.0	5000.0
30pc	29.0	114.3	1482.8	309.7	5000.0	3591.0	3431.0	1803.0	5000.0	5000.0	5000.0
50pc	22.0	88.9	1232.0	236.6	5000.0	3052.7	2850.0	1420.0	5000.0	5000.0	5000.0
70pc	19.5	77.7	1104.0	203.7	5000.0	2785.9	2521.0	1240.1	5000.0	5000.0	5000.0
90pc	17.0	71.5	1030.0	183.7	5000.0	2582.6	2372.0	1159.0	5000.0	5000.0	5000.0

^{240}Pu

This isotope of plutonium also had mass limits dictated by decay heat considerations for some of the weaker (α, n) light elements though to a lesser extent than ^{238}Pu. ^{240}Pu has a neutron source from spontaneous fissions that is an important contributor to the dose rates for the weaker (α, n) light elements. Table 4 presents the content envelope.

Table 4. ^{240}Pu Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	945.69	1913.58	2687.03	2489.43	2687.03	2687.03	2687.03	2687.03	2687.03	2687.03	2687.03
5000ppm	260.14	821.78	2635.11	1630.63	2687.03	2687.03	2687.03	2660.12	2687.03	2687.03	2687.03
10000ppm	138.48	486.47	2452.92	1144.40	2687.03	2687.03	2559.09	2416.46	2687.03	2687.03	2687.03
50000ppm	31.31	121.18	1480.99	353.53	2643.10	2182.29	1813.50	1522.38	2646.92	2687.03	2687.03
10pc	17.47	68.38	1059.22	204.55	2404.85	1810.94	1377.10	1082.87	2486.80	2622.05	2687.03
30pc	8.02	31.87	602.18	93.30	2024.80	1218.44	826.66	596.43	2121.93	2273.61	2500.80
50pc	6.18	24.75	486.60	69.98	1865.01	1011.35	663.16	463.98	1943.63	2127.24	2321.88
70pc	5.38	21.64	432.33	59.93	1772.82	904.31	586.69	405.29	1837.12	2044.80	2241.51
90pc	4.98	19.92	410.02	54.06	1705.51	842.44	540.36	369.62	1768.56	1991.12	2179.19

^{242}Pu

This isotope generally has a weaker dependence on light element (α, n) light element source of neutrons than ^{240}Pu. However, it has a stronger spontaneous fission source than ^{240}Pu since its half-life for this process is shorter than that of ^{240}Pu. Examining the content envelope for ^{242}Pu and beryllium in Table 5 with that for ^{240}Pu (see Table 4), it can be seen that the mass limits for the latter are smaller than for the former for each concentration. This is due to the fact ^{240}Pu has a higher source from (α, n) reactions in beryllium than ^{242}Pu. On the other hand, for weaker (α, n) sources from oxygen or chlorine, the spontaneous fission source tends to dominate, making the dependence on the light element content almost negligible. In these cases ^{240}Pu has higher mass limits than ^{242}Pu. Both these isotopes as well as ^{239}Pu have mass limits at the kilogram levels for combinations with most light elements barring beryllium, boron, fluorine, and, to a lesser extent, lithium.

Table 5. ^{242}Pu Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	1964.95	1989.97	2001.99	1999.15	2002.51	2001.66	2002.41	2002.39	2002.37	2002.38	2002.39
5000ppm	1827.12	1954.34	2001.19	1986.56	2002.30	2001.47	1999.97	2001.88	1999.66	2000.01	2002.37
10000ppm	1672.06	1908.40	2001.21	1973.13	2001.91	2000.79	2000.70	2000.01	1999.51	1999.60	2002.33
50000ppm	1070.12	1639.68	1995.09	1885.87	2000.65	1998.06	1995.13	1987.21	1998.20	1998.99	2001.95
10pc	783.49	1442.66	1988.24	1805.78	1998.76	1993.47	1986.87	1976.50	1997.08	1998.29	2001.71
30pc	460.39	1094.11	1971.55	1605.70	1995.10	1984.56	1970.58	1954.49	1993.95	1994.69	2000.82
50pc	372.18	963.75	1969.98	1503.98	1992.76	1978.17	1950.70	1942.72	1992.40	1993.44	2000.26
70pc	332.43	901.47	1964.50	1433.04	1991.48	1974.41	1941.47	1924.83	1990.71	1992.77	1999.76
90pc	309.07	858.50	1962.34	1396.96	1990.54	1973.68	1937.97	1925.34	1991.37	1992.55	2001.59

^{241}Am+^{241}Pu

Since ^{241}Pu beta decays with a half-life of 14.35 years to its daughter ^{241}Am and the latter is the dominant source of the external dose rates, it is more useful to present a combined content envelope for these two isotopes. The peak neutron source as a result of the decay of ^{241}Pu occurs in little over 73 years at which time 89% of the original ^{241}Pu has been converted to ^{241}Am. ^{241}Am combined with light elements is a good source of neutrons produced via the (α, n) reaction. Table 6 presents the envelope for this pair of actinides. In several instances the mass limits are restricted over all concentrations of light elements by decay heat considerations. Thus, these mass limits are conservative in terms of the external radiation levels they would produce.

Table 6. ^{241}Am+ ^{241}Pu Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	68.82	165.94	165.94	165.94	165.94	165.94	165.94	165.94	165.94	165.94	165.94
5000ppm	14.66	59.07	165.94	149.78	165.94	165.94	165.94	165.94	165.94	165.94	165.94
10000ppm	7.35	29.75	165.94	79.98	165.94	165.94	165.94	165.94	165.94	165.94	165.94
50000ppm	1.66	6.83	81.88	17.77	165.94	165.94	165.94	104.76	165.94	165.94	165.94
10pc	0.92	3.80	46.21	9.73	165.94	157.00	102.68	56.45	165.94	165.94	165.94
30pc	0.42	1.77	21.60	4.32	165.94	70.86	46.05	24.63	165.94	165.94	165.94
50pc	0.32	1.36	16.32	3.23	165.94	52.97	34.41	18.53	165.94	165.94	165.94
70pc	0.28	1.19	14.23	2.76	165.94	45.35	29.36	15.81	165.94	165.94	165.94
90pc	0.26	1.09	13.11	2.50	165.94	40.87	17.47	14.24	165.94	165.94	165.94

^{243}Am

This isotope is a strong gamma emitter and other than in combination with the larger concentrations of beryllium or boron, where the mass limit is 1 g, the mass limit remains constant at 1.32 g for every other light element. Table 7 presents the mass limits for this actinide.

Table 7. ^{243}Am Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
5000ppm	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
10000ppm	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
50000ppm	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
10pc	1.00	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
30pc	1.00	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
50pc	1.00	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
70pc	1.00	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32
90pc	1.00	1.00	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32	1.32

^{237}Np

This isotope is a predominantly a gamma emitter and other than for some higher concentrations of beryllium, the mass limits are constant as shown in Table 8.

Table 8. ^{237}Np Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
5000ppm	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
10000ppm	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
50000ppm	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
10pc	1005.79	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
30pc	1005.79	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
50pc	1005.79	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
70pc	1005.79	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62
90pc	1005.79	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62	2912.62

^{244}Cm

This isotope of curium has strong neutron sources from both spontaneous fission and (α, n) reactions with light elements. Thus the neutron dose rate is the dominant factor in contributing to external radiation. The mass limits across all light elements and concentrations are in the milligram range. Table 9 presents the mass limits.

Table 9. ^{244}Cm Mass limits in grams

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
1000ppm	0.23	0.25	0.26	0.25	0.26	0.26	0.26	0.26	0.26	0.26	0.26
5000ppm	0.17	0.23	0.25	0.25	0.26	0.26	0.26	0.25	0.26	0.26	0.26
10000ppm	0.13	0.21	0.25	0.23	0.26	0.25	0.25	0.25	0.26	0.26	0.26
50000ppm	0.05	0.13	0.23	0.18	0.25	0.25	0.25	0.23	0.26	0.26	0.26
10pc	0.03	0.09	0.21	0.14	0.25	0.24	0.24	0.22	0.25	0.26	0.25
30pc	0.01	0.05	0.17	0.09	0.25	0.23	0.21	0.18	0.25	0.25	0.25
50pc	0.01	0.04	0.15	0.07	0.25	0.22	0.20	0.17	0.25	0.25	0.24
70pc	0.01	0.04	0.15	0.06	0.25	0.21	0.20	0.16	0.25	0.25	0.24
90pc	0.01	0.03	0.14	0.06	0.25	0.21	0.19	0.15	0.24	0.25	0.24

CONCLUSIONS

This study has developed a set of conservative mass limits for the Model 9977 radioactive material transportation package that are compliant with both regulatory external radiation limits as well as design decay heat limits while considering the impact of subcritical multiplication in these actinides. It is noted that the limits incorporate margins ranging from 7.5% to 2.5% to the regulatory limits for the package surface dose rate, which was the limiting parameter. In addition, the model used is also very conservative adding further margins to the regulatory limits.

The Model 9977 Packaging has a polyethylene shielded container that has been approved for use with neutron emitting actinides. References 1 and 3 also present mass limits for the Model 9977 Packaging with the polyethylene shielded container. This study did not reevaluate the mass limits presented in Reference 3 when the polyethylene shielded container is included in the packaging. However, based on the present study with unshielded actinide and light element contents in the packaging, it is not unreasonable to expect that even accounting for subcritical multiplication, the mass limits from Reference 3 will yield low external radiation levels with large margins to the regulatory limits. This will be especially the case for those limits that were already producing very low external radiation levels in the unshielded case. It is noted that the decay heat limit for the polyethylene shielded container is 3 W and the mass limits presented in Reference 3 account for this more restrictive limit.

For mixtures of isotopes, the sum of the fraction of each isotope to its individual limit should be less than or equal to 1 for compliance with the regulatory limits, i.e.

$$\sum_{i=1}^{n} \frac{M_i}{M_{Li}} \leq 1$$

where,

M_i is the mass of the i^{th} isotope in the mixture containing n isotopes, and

M_{Li} is the maximum allowed mass for that isotope with the appropriate impurity fraction.

The content envelope results for the Model 9977 can be applied to and are conservative for the Model 9975 and the Model 9978 Packagings. The Model 9975 Packaging has double containment (inner and outer containment vessels) and a gamma shield. The Model 9978 Packaging has a 5-inch containment vessel compared to a 6-inch containment vessel for the Model 9977 Packaging, making the source to dose measurement point distance larger.

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